1	Effectiveness of precursor emission reductions for the control of summertime ozone
2	and $PM_{2.5}$ in the Beijing–Tianjin–Hebei region under different meteorological
3	conditions
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10	ABSTRACT
11	We used the observed concentrations of air pollutants, reanalyzed meteorological
12	parameters, and the results from the Goddard Earth Observing System Chemical
13	Transport Model (GEOS-Chem) to examine the relationships between maximum daily 8
14	h average ozone (MDA8 $O_3$ ), PM <sub>2.5</sub> (particulate matter with diameter of 2.5 $\mu$ m or less),
15	$PM_{2.5}$ components and 2 m temperature (T2)/relative humidity (RH) as well as the
16	effectiveness of precursor emission reductions on the control of O <sub>3</sub> and PM <sub>2.5</sub> in Beijing-
17	Tianjin-Hebei (BTH) under different summertime temperature and humidity conditions.
18	Both observed (simulated) MDA8 O <sub>3</sub> and PM <sub>2.5</sub> concentrations increased as T2 went up,
19	with linear trends of 4.8 (3.2) ppb $^{\circ}C^{-1}$ and 1.9 (1.5) $\mu g~m^{-3}~^{\circ}C^{-1},$ respectively. Model
20	results showed that the decreases in MDA8 O <sub>3</sub> from precursor emission reductions were
21	more sensitive to T2 than to RH. Reducing a larger proportion of volatile organic
22	compounds (VOCs) emissions at higher T2 was more effective for the control of

\*Corresponding author: Hong LIAO Email: <u>hongliao@nuist.edu.cn</u> summertime  $O_3$  in BTH. For the control of summertime  $PM_{2.5}$  in BTH, nitrogen oxides (NO<sub>x</sub>) reduction combined with a small proportion reduction of VOCs was the best measure. The magnitude of reduction in  $PM_{2.5}$  from reducing precursor emissions was more sensitive to RH than to T2, with the best efficiency at high RH. Results from this study are helpful for formulating effective policies to tackle  $O_3$  and  $PM_{2.5}$  pollution in BTH.

29 Key words: Ozone; PM<sub>2.5</sub>; Emission reductions; Meteorological conditions

30 Article Highlights:

- Observed summertime concentrations of O<sub>3</sub> and PM<sub>2.5</sub> in BTH increased as T2 and
   RH increased (when RH < 60%).</li>
- Larger percentage reduction of VOCs emissions at higher temperature is more
   effective for alleviating summertime O<sub>3</sub> pollution in BTH.
- NO<sub>x</sub> reduction combined with a small proportion reduction of VOCs is the best
   measure for PM<sub>2.5</sub>, which has the best effect at high RH.
- For the co-pollution of O<sub>3</sub> and PM<sub>2.5</sub> in summer in BTH, emission reductions have
  the best effect in a hot and humid conditions.
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### 42 1. Introduction

43 China has high concentrations of both PM<sub>2.5</sub> (particulate matter with an aerodynamic equivalent diameter of 2.5  $\mu$ m or less) and tropospheric ozone (O<sub>3</sub>), especially in the 44 Beijing-Tianjin-Hebei (BTH) region, a major urban cluster with developed economies in 45 eastern China (Luo et al., 2022; Dai et al., 2023). Over 2013-2019, the summer (June-46 July–August, JJA) mean maximum daily 8 h average (MDA8) O<sub>3</sub> concentrations were 47 observed to increase at a rate of 3.3 ppb  $y^{-1}$  over the North China Plain (NCP) (Li et al., 48 2020). In spite of the effective control of PM2.5 pollution in China since the 49 implementation of Clean Air Action in 2013, the annual mean PM2.5 concentration 50  $m^{-3}$ 51 averaged the BTH 44 in 2022 over μg was (https://www.mee.gov.cn/hjzl/sthjzk/zghjzkgb/), which still far exceeded the air quality 52 standard of 5  $\mu$ g m<sup>-3</sup> set by the World Health Organization. Further efforts are needed to 53 54 control O<sub>3</sub> and PM<sub>2.5</sub> pollution in BTH (Zhao et al., 2021).

Variations of O<sub>3</sub> and PM<sub>2.5</sub> are highly sensitive to changes in meteorology. While 55 wind, precipitation, and planetary boundary layer height may influence the dispersion, 56 wet removal, and vertical mixing of air pollutants (Chen et al., 2020; Zhang and Wang, 57 2020), temperature and relative humidity (RH) play important roles in the formation of 58 secondary pollutants and the gas-particle partitioning of semi-volatile components (Wang 59 et al., 2013; Shah et al., 2018; Leung et al., 2020). Higher temperature can accelerate the 60 61 chemical production of O<sub>3</sub>, while higher RH increases the O<sub>3</sub> loss (Johnson et al., 1999; 62 Jacob and Winner, 2009). As for PM<sub>2.5</sub>, higher temperature can enhance gas phase reaction rates and oxidant concentrations, leading to higher sulfate  $(SO_4^2)$  concentrations 63 64 (Liao et al., 2006). However, higher temperature can promote the volatilization of ammonium nitrate (Kleeman, 2008). Higher RH is conducive to aqueous phase aerosol 65

chemistry for the formation of secondary PM<sub>2.5</sub> (Dawson et al., 2007; Zheng et al., 2015). 66 67 Li et al. (2019a) showed by a stepwise multiple linear regression (MLR) model that the 68 changes in daily maximum 2 m temperature (Tmax), 10 m meridional wind, and RH explained 66% of monthly variability in summer MDA8 O<sub>3</sub> in BTH over 2013–2017. By 69 70 applying the Goddard Earth Observing System Chemical Transport Model 71 (GEOS-Chem) and the Lindeman, Merenda, and Gold (LMG) method, Dang et al. (2021) found that the changes in Tmax and RH explained, respectively, 31% and 11% of the 72 73 simulated interannual variations of MDA8 O<sub>3</sub> in JJA of 2012–2017 over NCP in the 74 simulation with changes in meteorological parameters alone. Zong et al. (2021) analyzed the synoptic weather pattern corresponding to the cooccurrence of O<sub>3</sub> and PM<sub>2.5</sub> pollution 75 during summer of 2015-2018 over eastern China by applying the T-mode principal 76 77 component analysis, and found that the warm and moist flow brought by the western Pacific subtropical high promoted hygroscopic growth of the fine particulate matter, 78 resulting in the increases in PM2.5 concentrations in BTH. 79

80 Emission reduction is the primary measure for controlling air pollution. Owing to the absence of effective emission control measures on volatile organic compounds (VOCs) in 81 previous policies in China, anthropogenic VOCs emissions in China were estimated to 82 increase from 25.9 Tg in 2010 to 28.6 Tg in 2017 (increase by 11%) according to the 83 84 Multi-resolution Emission Inventory for China (Zheng et al., 2018). In the 14th 85 Five-Year Plan for 2021–2025, China identifies VOCs emission management as a prioritization in order to further improve air pollution. In addition, as a common and 86 important precursor of  $O_3$  and  $PM_{2.5}$ , the continuous control of nitrogen oxides (NO<sub>x</sub> = 87 88  $NO + NO_2$ ) emissions remains a key focus of air pollution control. A number of studies

89 have examined VOCs and  $NO_x$  emission reduction strategies for the synergistic reduction 90 of ambient O<sub>3</sub> and PM<sub>2.5</sub> pollution. By using simulations from the GEOS-Chem for summer of 2017, Li et al (2019b) evaluated the effects of the reduction targets for 2018-91 2020 (i.e., 9% for NO<sub>x</sub> emissions, 10% for VOCs emissions and 8% for  $PM_{2.5}$ 92 concentrations) over NCP, and showed that the increases in O<sub>3</sub> owing to the decreases in 93 PM<sub>2.5</sub> were offset by the decreases in O<sub>3</sub> owing to the decreases in NO<sub>x</sub> and VOC 94 emissions, resulting in the reduction of about 1 ppb in seasonal mean MDA8 O<sub>3</sub>. Xiang et 95 96 al. (2020), by applying Weather Research and Forecasting model (WRF) with Chemistry 97 together with an empirical kinetics modeling approach to conduct multiple scenario analyses for reduction of anthropogenic NO<sub>x</sub> and VOCs emissions over the BTH and 98 surrounding areas relative to January and July of 2015–2017, and suggested that, from a 99 long-term perspective, reducing firstly VOCs emissions by about 60% and NO<sub>x</sub> 100 emissions by about 20% to avoid the rebound of O<sub>3</sub> and then phasing out the remaining 101 emissions to reach a deep mitigation. Ding et al. (2022) adopted the Community 102 103 Multiscale Air Quality (CMAQ) model, WRF model, and the extended response surface model with polynomial functions, and reported that no matter how much VOCs 104 emissions was reduced,  $NO_x$  emission reduction (64%–81%) was essential to attain the 105 air quality standard (with the annual PM<sub>2.5</sub> concentration  $\leq$  35 µg m<sup>-3</sup> and the annual 90th 106 percentile MDA8 O<sub>3</sub> concentration  $\leq 160 \ \mu g \ m^{-3}$ ) in BTH in 2017, in which year the 107 observed annual concentrations of  $O_3$  and  $PM_{2.5}$  were 178-216 µg m<sup>-3</sup> and 57-86 µg 108  $m^{-3}$ , respectively. These studies with the impacts of VOCs and NO<sub>x</sub> emission reductions 109 on O3 and PM2.5 were generally based on the average over time scales (i.e., monthly, 110 111 seasonal, annual), ignoring the impacts of variations in meteorological fields on control

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measures. Therefore, it is essential to examine the effectiveness of precursor emission reductions for the control of  $O_3$  and  $PM_{2.5}$  pollution under different conditions of temperature and humidity so as to formulate more effective policies.

The objectives of this study are (1) to examine the relationships between 115 116 concentrations of O<sub>3</sub> and PM<sub>2.5</sub> and T2/RH based on the observations from Chinese 117 Ministry of Ecology and Environment (MEE) and the reanalyzed meteorological parameters from Version 2 of Modern-Era Retrospective analysis for Research and 118 119 Application (MERRA-2) over BTH for summer of 2019, (2) to apply the GEOS-Chem 120 model that can capture the observed relationships to quantify the effectiveness of emission reductions of VOCs and NOx on O3 and PM2.5 in BTH under different 121 conditions of temperature and humidity, and (3) to identify the temperature and humidity 122 123 conditions under which emission reductions have the best effect for alleviating summertime O<sub>3</sub> and PM<sub>2.5</sub> pollution in BTH. Year 2019 was selected because MDA8 O<sub>3</sub> 124 concentrations in this year were among the highest in recent years, and the concentrations 125 of  $PM_{2.5}$  in 2019 were still at a relatively high level in BTH. 126

The remainder of this paper is structured as follows. Section 2 describes the observations, reanalyzed meteorological data, GEOS-Chem model, and numerical experiments. The model evaluation, the relationships between atmospheric oxidants,  $O_3$ , PM<sub>2.5</sub>, and PM<sub>2.5</sub> components and T2/RH, as well as the simulated changes of all these species under different temperature and humidity conditions with emission reductions of VOCs and/or NO<sub>x</sub> are presented in Section 3. Section 4 summarizes the main conclusions.

### 134 **2. Methods**

#### 135 2.1 Observed $O_3$ and $PM_{2.5}$ concentrations

The observed hourly concentrations of O<sub>3</sub> and PM<sub>2.5</sub> for JJA of year 2019 were 136 obtained from MEE and can be downloaded from https://quotsoft.net/air/ (Wang, 2024). 137 There are over 1600 observational sites throughout the country, in which 79 are located 138 in Beijing-Tianjin-Hebei (BTH) region (37-41°N, 114-118°E). Concentrations were 139 140 reported by the MEE in micrograms per cubic meter ( $\mu g m^{-3}$ ) under reference state (298) K, 1013 hPa) for O<sub>3</sub> and under local ambient state for PM<sub>2.5</sub>. For the consistency between 141 observed and simulated O<sub>3</sub> concentrations, we converted the unit of observed O<sub>3</sub> to parts 142 143 per billion (ppb). For O<sub>3</sub>, MDA8 was used in this study, which was calculated when there were valid hourly data for at least 6 h for each 8 h and there were more than 14 valid 8 h 144 averaged data in each day. Daily mean PM<sub>2.5</sub> concentration was calculated when there 145 were valid hourly data for more than 20 h during that day. All the site concentrations 146 were averaged within each of the  $0.5^{\circ}$  latitude  $\times 0.625^{\circ}$  longitude grid cells of MERRA-2 147 for model evaluation and subsequent analysis, following Dai et al. (2023). There are 18 148 149 model grids in BTH. We consider each day of each grid cell as an independent sample.

### 150 2.2 Reanalyzed meteorological data

Meteorological fields for 2019 were obtained from MERRA-2 generated by the NASA Global Modeling and Assimilation Office (GMAO). The MERRA-2 data have a horizontal resolution of  $0.5^{\circ}$  latitude  $\times 0.625^{\circ}$  longitude. Meteorological parameters considered in the study include 2 m temperature (T2), RH, and precipitation (PR). The temporal resolution for T2 and PR is 1 h, and that for RH is 3 h. Note that T2 and RH are averaged over 24 h, while PR is summed over 24 h. We do not consider days with precipitation, samples with total daily precipitation greater than 0.1 mm were removed. Meanwhile, after removing the days with missing data of MDA8  $O_3$  or  $PM_{2.5}$ , there are ultimately 620 valid samples remaining out of 1656 samples over BTH in JJA of 2019. To explore the effects of precursor emission reductions on MDA8  $O_3$  and  $PM_{2.5}$  under different conditions of temperature and humidity, T2 are binned to 2°C intervals and RH are binned to 5% intervals.

#### 163 2.3 GEOS-Chem model

The simulations of air pollutants for JJA of year 2019 were carried out by using the 164 nested version of 165 **GEOS-Chem** (version 11-01. http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem v11-01), driven by the 166 MERRA-2 assimilated meteorological fields (Gelaro et al., 2017). The nested version of 167 v11-01 in the Asian (11°S–55°N, 60°E–150°E) has a horizontal resolution of 0.5° latitude 168  $\times$  0.625° longitude and 47 vertical layers up to 0.01 hPa, with dynamical boundary 169 conditions from a global GEOS-Chem simulation by  $2^{\circ}$  latitude  $\times 2.5^{\circ}$  longitude 170 171 horizontal resolution. The O<sub>3</sub> and PM<sub>2.5</sub> simulation in the GEOS-Chem model has been extensively used in previous studies (Fu and Liao, 2014; Zhang et al., 2018; Wang and 172 Liao, 2020; Gong et al., 2022). The GEOS-Chem model includes a fully coupled 173 174 O<sub>3</sub>-NO<sub>x</sub>-hydrocarbon chemistry (Bey et al., 2001; Park et al., 2004) and aerosols including sulfate (Park et al., 2004), nitrate (NO $_3$ ) (Pye et al., 2009), ammonium (NH $_4^+$ ), 175 176 black carbon (BC) and organic carbon (OC) (Park et al., 2003), mineral dust (Fairlie et al., 2007), and sea salt (Alexander et al., 2005). Two primary organic aerosol (POA) 177 178 tracers are considered in the model: hydrophobic and hydrophilic POAs. The 179 hydrophobic POA becomes hydrophilic with an e-folding time of 1.15 days (Cooke et al., 1999; Park et al., 2003). It is assumed that 50% of POAs emitted are hydrophobic (Park 180

181 et al., 2003). The simulation of secondary organic aerosol (SOA) is based on the volatility basis set (VBS) approach. Reactions of forming SOA from different parent 182 hydrocarbons can be mapped onto the same set of bins encompassing the range of typical 183 ambient organic aerosol mass concentrations ( $\sim 0.1-100 \ \mu g \ m^{-3}$ ) (Jo et al., 2013). Wet 184 deposition, including washout, rainout, and scavenging in moist convective updrafts, 185 follows the scheme of Liu et al. (2001). Dry deposition is calculated based on the 186 resistance-in-series model described by Wesely (1989) with a number of modifications 187 (Wang et al., 1998). 188

189 The anthropogenic emissions of NO<sub>x</sub>, carbon monoxide, OC, BC, sulfur dioxide (SO<sub>2</sub>), and ammonia (NH<sub>3</sub>) were obtained from MEIC (http://meicmodel.org.cn/) for year 190 2019, which includes emissions from the industry, power, residential, and transportation 191 sectors. Due to the selection of a chemical mechanism developed by the State Air 192 Pollution Research Center (SAPRC), namely SAPRC99, the downloaded MEIC data lack 193 aromatics species that have a significant impact on SOA formation, such as benzene and 194 195 toluene (Zhang et al., 2021; Wang et al., 2022). As a result, the anthropogenic emissions 196 of VOCs (including ethane, propane, butanes, pentanes, other alkanals, ethene, propene, acetylene, benzene, toluene, xylene, formaldehyde, total ketones, and total acids) were 197 Shared Socioeconomic 198 obtained from Pathway (SSP) emission inventory 199 (https://esgf-node.llnl.gov/projects/input4mips/). Year 2020 emissions of SSP2-4.5 were 200 used in our simulations, which represent a middle-of-the-road development in the mitigation and adaptation challenges space. The biogenic emissions in GEOS-Chem were 201 taken from MEGAN v2.1 (Guenther et al., 2012). 202

#### 203 2.4 Numerical experiments

To investigate the effects of VOCs emission reductions on  $O_3$  and  $PM_{2.5}$  in BTH under different meteorological conditions, four numerical experiments were performed for JJA of 2019. Considering the recent national policies and the feasibility of practical control measures, the reduction ratios of anthropogenic VOCs were set between 10% and 50% with a 20% interval:

- 209 (1) CTRL: The control simulation without emission control measures;
- 210 (2) VOCs\_10: The same as the CTRL simulation but with a 10% reduction in
  211 anthropogenic VOCs emissions in China;
- 212 (3) VOCs\_30: The same as the CTRL simulation but with a 30% reduction in
  213 anthropogenic VOCs emissions in China;
- (4) VOCs\_50: The same as the CTRL simulation but with a 50% reduction in
  anthropogenic VOCs emissions in China;
- Since  $NO_x$  also plays an important role in the formation of  $O_3$  and  $PM_{2.5}$ , and the
- new 14th Five-Year Plan for 2021–2025 calling for a 10% decrease in NO<sub>x</sub> emissions
- 218 (<u>https://www.mee.gov.cn/zcwj/zyygwj/202111/t20211108\_959456.shtml</u>), we also
- 219 conducted three additional experiments:
- (5) NO<sub>x</sub>\_10+VOCs\_10: The same as the CTRL simulation but with a 10% reduction
  in both anthropogenic NO<sub>x</sub> and anthropogenic VOCs emissions in China;
- 222 (6) NO<sub>x</sub>\_10+VOCs\_30: The same as the CTRL simulation but with 10% and 30%
- reductions in anthropogenic NO<sub>x</sub> and VOCs emissions in China, respectively;
- 224 (7) NO<sub>x</sub>\_10+VOCs\_50: The same as the CTRL simulation but with 10% and 50%
- reductions in anthropogenic  $NO_x$  and VOCs emissions in China, respectively;

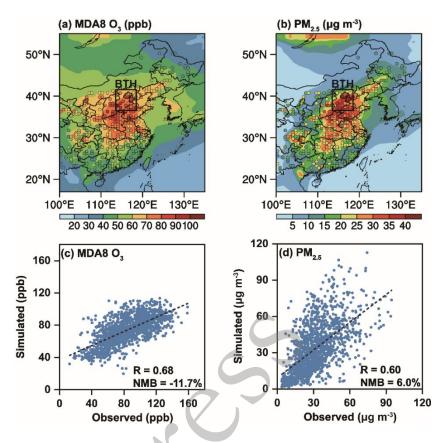
All the simulations were integrated for the period of 1 June to 31 August of year 2019 after a six month spin up of the model.

228 **3. Results** 

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# 3.1 Evaluation of model performance

230 Figures 1a and 1b present, respectively, the spatial distributions of simulated and 231 observed surface-layer concentrations of MDA8 O<sub>3</sub> and PM<sub>2.5</sub> averaged over summer of 232 2019. For model evaluation, observed MDA8 O<sub>3</sub> and PM<sub>2.5</sub> concentrations were averaged 233 over all sites within each of the  $0.5^{\circ} \times 0.625^{\circ}$  MERRA-2 grid cells. The concentrations of MDA8 O<sub>3</sub> and PM<sub>2.5</sub> were both relatively high in BTH. The seasonal mean observed 234 (simulated) concentrations of MDA8  $O_3$  and  $PM_{2.5}$  were 83.6 (71.9) ppb and 31.6 (32.1) 235 236  $\mu$ g m<sup>-3</sup>, respectively, as values were averaged over the 18 grids in BTH. Figures 1c and 1d show the scatterplot of simulated versus observed daily MDA8 O<sub>3</sub> and PM<sub>2.5</sub> 237 concentrations, respectively, for 18 grids located in BTH. The simulated spatiotemporal 238 variations of MDA8 O<sub>3</sub> and PM<sub>2.5</sub> agree fairly well with the observations, with 239 correlation coefficients (R) of 0.68 and 0.60, respectively. The model underestimates 240 MDA8  $O_3$  concentrations with a normalized mean bias (NMB) of -11.7% and 241 242 overestimates  $PM_{2.5}$  concentrations with a NMB of +6.0% in BTH.

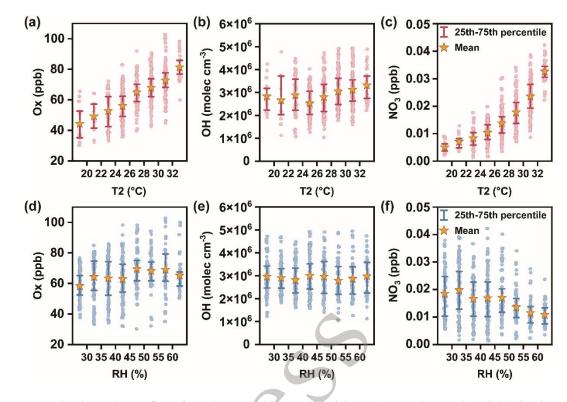


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Fig. 1. Spatial distributions of simulated (shades) and observed (dots) surface-layer 244 concentrations of (a) MDA8 O<sub>3</sub> (units: ppb) and (b) PM<sub>2.5</sub> (units: µg m<sup>-3</sup>) averaged over 245 summer in 2019. The solid black rectangles in (a-b) indicate the BTH region, which 246 contain 18 grids. Scatterplot of simulated versus observed daily concentrations of (c) 247 MDA8 O<sub>3</sub> (units: ppb) and (d) PM<sub>2.5</sub> (units:  $\mu$ g m<sup>-3</sup>) for 18 grids located in BTH. The 248 correlation coefficient (R) and normalized mean bias (NMB; units: %) are also shown in 249 250 (c-d). Here, R is the correlation coefficient between simulated and observed concentrations. NMB =  $(\sum_{i=1}^{N} (M_i - O_i)) / \sum_{i=1}^{N} O_i \times 100\%$ , where  $O_i$  and  $M_i$  are the 251 observed and simulated concentrations, respectively; *i* refers to the *i*th sample, and *N* 252 253 is the total number of samples.

#### 254 3.2 The relationships between atmospheric oxidants and T2/RH

255 The concentrations of total oxidant ( $O_x = O_3 + NO_2$ ), hydroxyl radical (OH), and nitrogen trioxide (NO<sub>3</sub>) are usually used to characterize the atmospheric oxidation 256 capacity (Kang et al., 2021; Tang et al., 2022). Figures 2a-c and 2d-f show the boxplots 257 of simulated daily concentrations of O<sub>x</sub>, OH, and NO<sub>3</sub> in T2 bins and in RH bins 258 (described in Sect. 2.2), respectively, considering the 18 grids of BTH in JJA of year 259 260 2019 (620 valid samples as also described in Sect. 2.2) from the CTRL simulation to investigate the levels of atmospheric oxidation capacity under different temperature and 261 humidity conditions. The O<sub>x</sub> and NO<sub>3</sub> concentrations gradually increased as T2 went up, 262 with the mean values increasing from 44.2 ppb and  $5.0 \times 10^{-3}$  ppb in the bin of  $\leq 20^{\circ}$ C to 263 81.3 ppb and  $3.3 \times 10^{-2}$  ppb in the bin of >32°C, respectively. The concentrations of OH 264 exhibited continuous increases as T2 was above 24°C, and the mean value increased from 265 2.5  $\times$  10<sup>6</sup> molec. cm<sup>-3</sup> at T2 of 24–26°C to 3.3  $\times$  10<sup>6</sup> molec. cm<sup>-3</sup> at >32°C. The 266 relationships between  $O_x/OH/NO_3$  and T2 indicate that the atmospheric oxidation 267 capacity is stronger at higher temperature, which favors the formation of both secondary 268 inorganic and organic particulates (Feng et al., 2019). As for the relationships between 269 atmospheric oxidation capacity and RH, there was a small increasing trend in simulated 270 271  $O_x$  concentrations as RH increased, with a peak value of 69.5 ppb as RH was 45–50%. 272 OH concentrations showed a W-shaped trend, with valleys occurring when RH values 273 were 35-40% and 50-55%. NO<sub>3</sub> concentrations had an overall decreasing trend as RH increased. 274



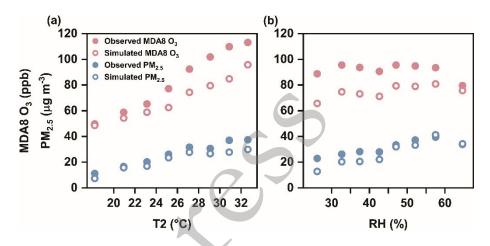
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Fig. 2. The boxplots of surface-layer (a/d) total oxidant ( $O_x$ ; units: ppb), (b/e) hydroxyl radical (OH; units: molec. cm<sup>-3</sup>), and (c/f) nitrogen trioxide (NO<sub>3</sub>; units: ppb) in each T2/RH bin in 18 grids of BTH in JJA of year 2019 (620 valid samples) from the CTRL simulation. T2 are binned to 2°C intervals and RH are binned to 5% intervals.

## 280 3.3 The relationships between air pollutants and T2/RH

281 3.3.1 The relationships between MDA8 O<sub>3</sub>/PM<sub>2.5</sub> and T2/RH

Figure 3 displays the relationships between observed and simulated concentrations of MDA8 O<sub>3</sub> and T2/RH in BTH during JJA in 2019 (620 valid samples as described in Sect. 2.2). Both observed and simulated MDA8 O<sub>3</sub> increased as T2 went up, with increasing rates of 4.8 ppb  $^{\circ}C^{-1}$  and 3.2 ppb  $^{\circ}C^{-1}$ , respectively. As RH increased, there was a bimodal feature in observed and simulated MDA8 O<sub>3</sub> concentrations, with the peaks appearing as RH values were 30–35% and 45–50% for observations (30–35% and 288 55–60% for model results). Both observed and simulated MDA8  $O_3$  concentrations 289 decreased as RH was greater than 60%. Compared to 55–60%, the net chemical 290 production of  $O_3$  had smaller positive values in the lower troposphere at RH>60% (Fig. 291 6c). In addition, the concentration of BC (a chemically inert species) also showed a drop 292 as RH was above 60% (Fig. 4j), indicating that physical processes also contributed to the 293 decrease in pollutant concentrations.



**Fig. 3.** Relationships between observed (solid dots) and simulated (hollow dots) MDA8 O<sub>3</sub> (pink dots)/PM<sub>2.5</sub> (blue dots) and (a) T2/(b) RH in 18 grids of BTH in JJA of 2019 (620 valid samples). Dots represent the mean of the daily concentrations of MDA8 O<sub>3</sub> (units: ppb) and PM<sub>2.5</sub> (units:  $\mu$ g m<sup>-3</sup>) (a) versus daily T2 (units: °C) in each T2 bin and (b) versus daily RH (units: %) in each RH bin. T2 are binned to 2°C intervals and RH are binned to 5% intervals.

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The relationships between observed and simulated concentrations of  $PM_{2.5}$  and T2/RH in BTH during JJA in 2019 were also shown in Fig. 3. On the whole, concentrations of observed (simulated)  $PM_{2.5}$  also increased as T2 rose, with linear trends of 1.9 (1.5) µg m<sup>-3</sup> °C<sup>-1</sup>.  $PM_{2.5}$  concentrations exhibited fast increases as T2 was between  $\leq 20^{\circ}$ C and 26–28°C but had a small drop as T2 was between 28–30°C and remained relatively stable as T2 was >30°C. As RH increased from  $\leq$ 30% to 55–60%, observed and simulated PM<sub>2.5</sub> concentrations continuously increased. Same as MDA8 O<sub>3</sub>, PM<sub>2.5</sub> concentrations decreased significantly as RH was above 60%. At 1005–913 hPa, net chemical productions of PM<sub>2.5</sub> had larger values at RH>60% than those in the previous humidity interval (Fig. 6d), so the physical processes dominated the drop of PM<sub>2.5</sub> at RH>60%, as again indicated by the drop of BC concentration. The model generally captures the variations in the observed MDA8 O<sub>3</sub>/PM<sub>2.5</sub> with T2/RH.

### 313 3.3.2 The relationships between $PM_{2.5}$ components and T2/RH

Figures 4a-e and 4f-i show the simulated surface concentrations of PM<sub>2.5</sub> 314 components (BC,  $SO_4^2$ ,  $NO_3^2$ ,  $NH_4^+$ , and SOA) and the vertical distributions of 315 concentrations of  $SO_4^{2-}$ ,  $NO_3^{-}$ ,  $NH_4^{+}$ , and total nitrate ( $NO_3^{T} = HNO_3 + NO_3^{-}$ ), 316 respectively, in T2 bins over BTH during JJA of year 2019 from the CTRL simulation. 317 The concentrations are averaged over the 18 grids in BTH. As T2 increased, the mean 318 concentrations of BC had a general upward trend. Since BC is assumed to be chemically 319 inert in the model (Dang and Liao, 2019), changes in BC concentrations can represent the 320 changes in pollutant concentrations by physical processes. The relationship between BC 321 322 and T2 indicates that physical processes were generally more conducive to the accumulation of pollutants at higher temperature. To identify the contributions of 323 chemical processes to concentrations under different temperatures, the vertical profiles of 324 net chemical production of  $SO_4^{2-}$ ,  $NO_3^{-}$ , and  $NH_4^{+}$  in each T2 bin in 18 grids of BTH 325 326 during JJA of year 2019 are shown in Figs. 5a-c. In the lower troposphere (from the surface to about 700 hPa altitude), the chemical production of  $SO_4^{2-}$  gradually increased 327 328 when T2 increased (Fig. 5a), consistent with the trends of atmospheric oxidants (Figs.

329 2a-c). Therefore, both physical and chemical processes contributed to the increases in
 330 SO<sup>2</sup>/<sub>4</sub> - concentrations as T2 increased (Fig. 4b).

For the formation of total nitrate, NO<sub>2</sub> oxidation by OH by NO<sub>2</sub> + OH + M  $\rightarrow$ 331  $HNO_3 + M$  is the major pathway during daytime, and the major reactions at nighttime 332 are NO +  $O_3 \rightarrow NO_2$  +  $O_2$ , NO<sub>2</sub> +  $O_3 \rightarrow NO_3$  +  $O_2$ , NO<sub>2</sub> + NO<sub>3</sub> +  $M \rightarrow N_2O_5$  + M, 333 and  $N_2O_5 + H_2O \rightarrow 2HNO_3$  (Dong et al., 2014; Leung et al., 2020). Higher 334 concentrations of  $NO_3^T$  at higher temperature (Fig. 4i) also confirms the enhancement of 335 atmospheric oxidation capacity. However, the variation of  $NO_3^-$  concentrations with T2 336 differed from that of  $NO_3^T$ . The surface concentration of  $NO_3^T$  at T2 of 22–24°C was 337 lower than that at 20–22°C (Fig. 4c) due to the larger negative net chemical productions 338 from the surface to 913 hPa at 22-24°C (Fig. 5b). Similarly, relative to 26-28°C, the net 339 chemical production of  $NO_3^-$  had lager negative values from the surface to 913 hPa and 340 consequently surface NO<sub>3</sub> had lower concentration at T2 of 28–30°C. At 959–898 hPa, 341 the net chemical productions of  $NO_3^-$  were more negative both at 30–32°C and at >32°C 342 than those in the previous temperature interval, resulting in the decreases in  $NO_3^{-1}$ 343 concentrations as T2 increased. Overall, in the lower troposphere, the lower 344 345 concentrations of  $NO_3^-$  at higher T2 were attributed to the evaporation of  $NO_3^-$ . For  $NH_{4}^{+}$ , as a result of the larger negative net chemical productions from the surface to 913 346 hPa at 28-32°C (Fig. 5c), concentrations at 28-32°C were lower than those at 26-28°C 347 (Fig. 4d). 348

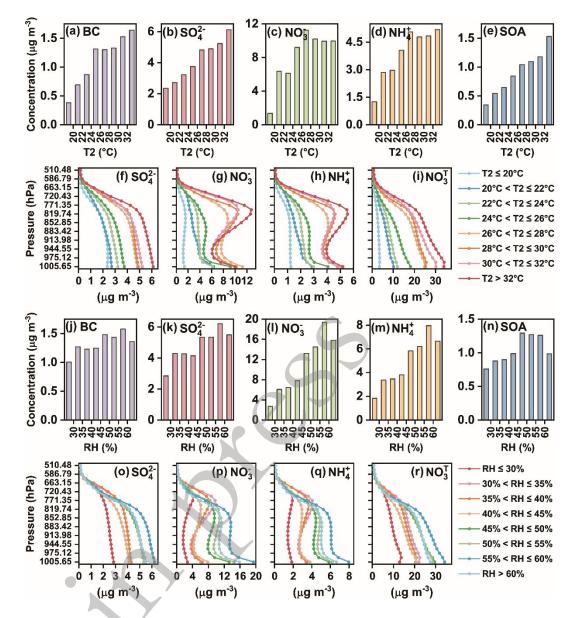


Fig. 4. (a–e/j–n) The surface concentrations (units:  $\mu g m^{-3}$ ) of PM<sub>2.5</sub> components (BC, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, and SOA) and (f–i/o–r) the vertical distributions of concentrations (units:  $\mu g m^{-3}$ ) of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, and total nitrate (NO<sub>3</sub><sup>T</sup> = HNO<sub>3</sub> + NO<sub>3</sub><sup>-</sup>) averaged over each T2/RH bin in 18 grids of BTH in JJA of year 2019 (620 valid samples) from the CTRL simulation.

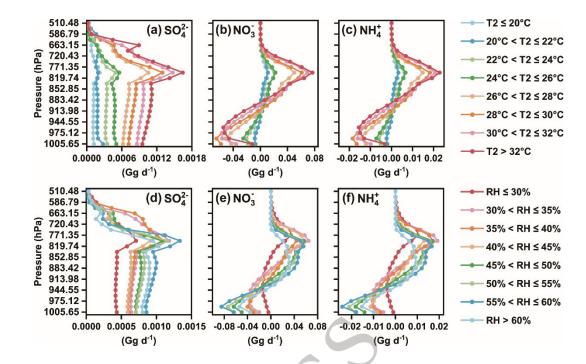


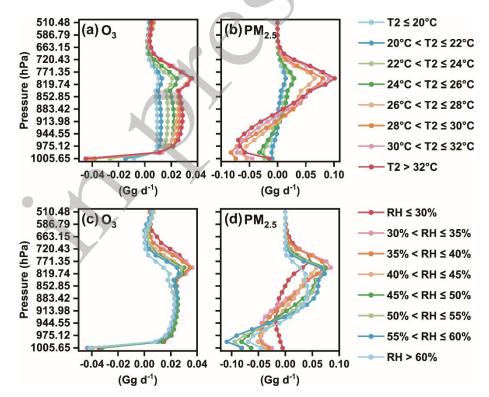
Fig. 5. The vertical profiles of net chemical production (units: Gg d<sup>-1</sup>) of (a/d)  $SO_4^{2^-}$ , (b/e) NO<sub>3</sub>, and (c/f) NH<sub>4</sub><sup>+</sup> in each T2/RH bin in 18 grids of BTH in JJA of year 2019 (620 valid samples) from the CTRL simulation.

High temperature promotes the evaporation of SOA and the emission of biogenic VOCs (Lee et al., 2011; Liu et al., 2019; Wu et al., 2020). The significant upward trend in SOA concentrations indicates the dominant roles of the elevated levels of biogenic VOCs emissions and the enhanced atmospheric oxidation capacity as T2 went up. Simulated SOA concentrations were the lowest among all secondary aerosols, so it should not drive the trend of changes in  $PM_{2.5}$  with T2.

The simulated surface and vertical concentrations of  $PM_{2.5}$  components in RH bins in BTH during JJA of 2019 are presented in Figs. 4j–r, and the change in net chemical production of secondary inorganic components with RH is presented in Figs. 5d–f. As RH increased, the mean concentrations of BC showed a fluctuating upward trend and a large drop as RH was above 60%, with three peaks appearing at 30–35%, 45–50%, and

55–60% (Fig. 4j).  $SO_4^{2-}$  concentrations mimicked the variations in BC to some extent. 370 Different from the changes in  $SO_4^2$  concentrations, the concentrations of  $NO_3^2$ 371 gradually increased with RH when RH was below 60%, and the changes in  $NH_4^+$ 372 followed that of NO<sub>3</sub> (Figs. 41–m). For SOA, the concentration increased first and then 373 decreased as RH went up, with a peak concentration of 1.3  $\mu$ g m<sup>-3</sup> occurring when RH 374 was 45-50%. Consistent with BC, concentrations of PM<sub>2.5</sub> secondary components 375 376 showed a significant drop at RH of >60%, indicating the role of physical process. With respect to the role of chemical process, the net chemical production of  $SO_4^{2-}$  generally 377 increased with RH, whereas that of  $NO_3^-$  and  $NH_4^+$  had larger net loss at higher RH 378

379 (Figs. 5d-f).



**381** Fig. 6. Same as Fig. 5, but for  $(a/c) O_3$  and  $(b/d) PM_{2.5}$ .



Six numerical experiments (VOCs 10, VOCs 30, VOCs 50, NO<sub>x</sub> 10+VOCs 10, 384  $NO_x$  10+VOCs 30, and  $NO_x$  10+VOCs 50) were conducted to quantify the impacts of 385 different proportions of precursor emissions reduction in summer of 2019 on O<sub>3</sub> and 386 PM<sub>2.5</sub> in the BTH region under different temperature and humidity conditions. Figures 387 7a-d and 7e-h show the mean changes ( $\Delta$ ) of OH, O<sub>3</sub>, NO<sub>3</sub>, and sulfur oxidation ratio 388 (SOR =  $n - SO_4^2$  / ( $n - SO_4^2$  +  $n - SO_2$ ), where  $n - SO_4^2$  and  $n - SO_2$  are the molar 389 concentrations of  $SO_4^2$  and  $SO_2$ , respectively) in T2 bins and in RH bins, respectively, 390 relative to the CTRL simulation. SOR is used to quantify the conversion degrees of sulfur 391 (Zhu et al., 2019). 392

The main sources of OH radical in urban areas are produced by the photolysis of  $O_{3}$ , 393 formaldehyde (HCHO) and nitrous acid (HONO), and the main sink of OH is to react 394 with NO<sub>x</sub>, VOCs and peroxy radicals (HO<sub>2</sub>) (Li et al., 2021; Dai et al., 2024). 395 Concentrations of OH mostly increased in the sensitivity simulations with emissions 396 reductions relative to the CTRL simulation. Further investigation showed that in cases of 397 reducing VOCs emissions alone, the increase in OH concentration was mainly driven by 398 the enhancement of photolysis of HONO, while in cases of simultaneously reducing NO<sub>x</sub> 399 400 and VOCs emissions, it was mainly attributed to the weakening of the removal reaction with NO<sub>2</sub>. As T2 increased,  $\Delta OH$  concentrations peaked at 20–22°C in cases with 401 reductions of VOCs emissions alone and peaked at 24-26°C in cases with reductions of 402 403 both VOCs and  $NO_x$  emissions, with the largest reduction of  $NO_x$  in this interval (Fig. 13a).  $\Delta OH$  concentrations had a maximum value at RH of 45–50% except for the cases 404 of NO<sub>x</sub> 10+VOCs 10 and NO<sub>x</sub> 10+VOCs 30 simulations, which peaked at >60%.  $\Delta$ OH 405

406 was larger as the percentage reduction of VOCs was higher. Compared to the cases with 407 reductions of VOCs emissions alone, simultaneous reductions of  $NO_x$  and VOCs 408 promoted the formation of OH radical when T2 was above 24°C and in almost each RH 409 interval.

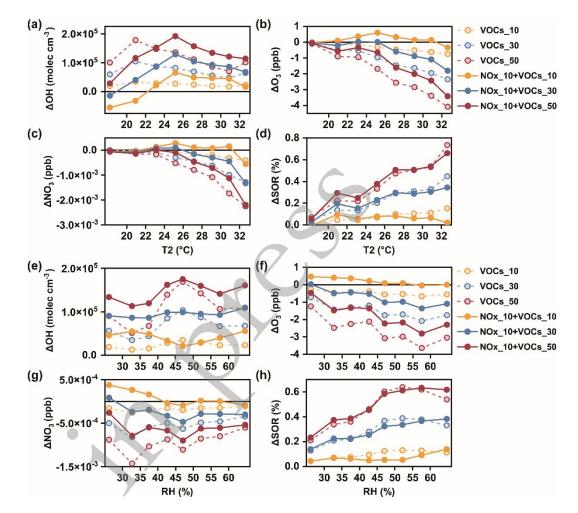


Fig. 7. Simulated mean changes in surface-layer (a/e) OH (units: molec. cm<sup>-3</sup>), (b/f) O<sub>3</sub> (units: ppb), (c/g) NO<sub>3</sub> (units: ppb), and (d/h) sulfur oxidation ratio (SOR; units: %) in each T2/RH bin in 18 grids of BTH in JJA of 2019 (620 valid samples) in simulations with reductions of anthropogenic VOCs emissions alone (dotted lines with hollow dots) and simulations with simultaneous reductions of anthropogenic NO<sub>x</sub> and VOCs emissions (solid lines with solid dots) relative to the CTRL simulation.

417 Concentrations of O<sub>3</sub> and NO<sub>3</sub> mostly decreased in the sensitivity simulations relative to the CTRL simulation.  $\Delta O_3$  and  $\Delta NO_3$  had larger negative values when T2 418 419 increased in all six simulations. When RH was lower than 60%,  $\Delta O_3$  had larger negative 420 values as RH increased. The highest reductions in NO<sub>3</sub> concentrations occurred at RH of 30-35% (45-50%) in cases with reductions of VOCs emissions alone (of both VOCs and 421 NO<sub>x</sub> emissions). The larger the percentage reduction of VOCs emissions, the higher the 422 423 decreases in O<sub>3</sub> and NO<sub>3</sub>. Reducing both NO<sub>x</sub> and VOCs emissions suppressed the decreases in O<sub>3</sub> and NO<sub>3</sub> concentrations relative to reducing VOCs emissions alone. 424

425 ΔSOR was generally positive in the sensitivity simulations relative to the CTRL 426 simulation. ΔSOR increased when T2 went up except for the case of NO<sub>x</sub>\_10+VOCs\_10, 427 in which the highest increase occurred at 20–22°C. The maximum ΔSOR occurred as RH 428 was 50–55% (above 55%) in cases with reductions of VOCs emissions alone (of both 429 NO<sub>x</sub> and VOCs emissions). The simulation with larger reduction of VOCs emissions led 430 to the higher increase in SOR.

# 431 3.4.2 Impact of precursor emission reductions on MDA8 O<sub>3</sub> concentrations

The absolute and percentage changes in MDA8 O<sub>3</sub> concentrations in each T2 and RH 432 intervals in BTH in JJA of 2019 with the six emission reduction measures are 433 434 summarized in Fig. 8. The changes in mean MDA8 O<sub>3</sub> concentrations were always negative. As T2 went up, the absolute values of  $\Delta$ MDA8 O<sub>3</sub> gradually increased, with 435 436 maximum values of 0.9, 2.7, 4.7, 3.4, and 5.2 ppb in simulations of VOCs 10, VOCs 30, VOCs 50, NO<sub>x</sub> 10+VOCs 30, and NO<sub>x</sub> 10+VOCs 50, respectively. In the 437 438 NO<sub>x</sub> 10+VOCs 10 simulation, although the decreases in MDA8 O<sub>3</sub> concentrations did 439 not increase monotonically with T2, the decreases in the 75th percentile of MDA8 O<sub>3</sub> did

440	(Fig. 9). As for the percentage changes in MDA8 $O_3$ , $NO_x_{10}+VOCs_{10}$ ,
441	NO <sub>x</sub> _10+VOCs_30, and NO <sub>x</sub> _10+VOCs_50 simulations had the lowest values (-1.7%,
442	-3.5%, and -5.4%, respectively) at T2 of >32°C, while VOCs_10, VOCs_30, and
443	VOCs_50 simulations exhibited the lowest values ( $-0.9\%$ , $-2.8\%$ , and $-4.9\%$ ,
444	respectively) at 30-32°C. When RH increased, the absolute and percentage decreases of
445	MDA8 O <sub>3</sub> concentrations both exhibited a bimodal trend, with the peaks appearing at RH
446	of 30–35% and 45–50% in the $\rm NO_x\_10+\rm VOCs\_10$ simulation and at 30–35% and
447	55-60% in other simulations. The highest reductions were simulated to be 0.8 (1.0%),
448	2.6 (3.1%), 4.6 (5.4%), 1.0 (1.2%), 2.7 (3.1%), and 4.5 ppb (5.2%) in VOCs_10,
449	VOCs_30, VOCs_50, NO <sub>x</sub> _10+VOCs_10, NO <sub>x</sub> _10+VOCs_30, and NO <sub>x</sub> _10+VOCs_50
450	simulations, respectively.

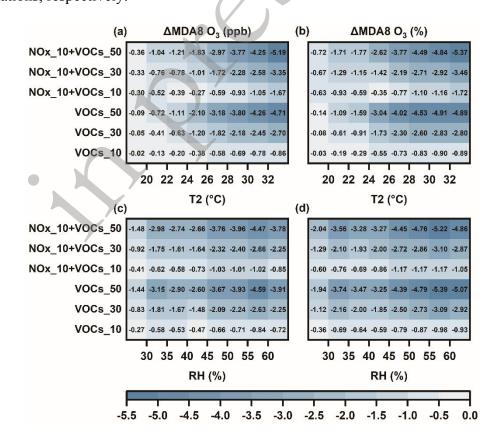
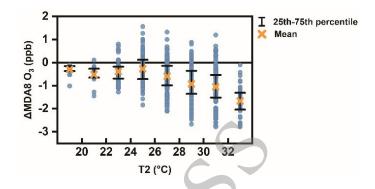


Fig. 8. (a)/(c) Absolute (units: ppb) and (b)/(d) percentage changes (units: %) in MDA8 O<sub>3</sub> concentrations in each T2 and bins in 18 grids of BTH in JJA of 2019 (620 valid samples) in VOCs\_10, VOCs\_30, VOCs\_50, NO<sub>x</sub>\_10+VOCs\_10, NO<sub>x</sub>\_10+VOCs\_30, and NO<sub>x</sub>\_10+VOCs\_50 simulations (from bottom to top) relative to the CTRL simulation.



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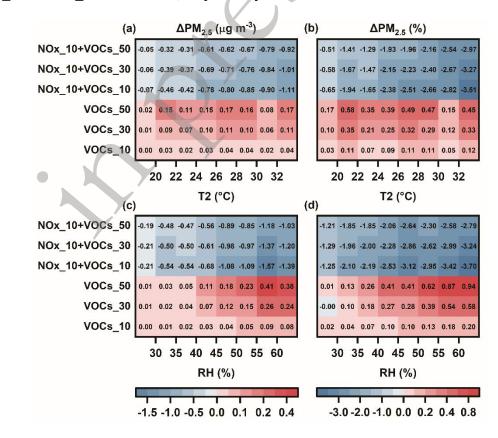
Fig. 9. The boxplots of absolute changes (units: ppb) in MDA8 O<sub>3</sub> concentrations in each
T2 bin in 18 grids of BTH in JJA of 2019 for all valid samples in the NO<sub>x</sub>\_10+VOCs\_10
simulation relative to the CTRL simulation.

In summary, implementing precursor emission reductions can effectively mitigate summer  $O_3$  pollution in BTH, especially in the case with the higher reduction of VOCs emissions. Furthermore, relative to the situations in RH intervals, a larger reduction of VOCs emissions at higher T2 (>30°C) showed better effectiveness of controlling  $O_3$ pollution.

## 466 3.4.3 Impact of precursor emission reductions on $PM_{2.5}$ concentrations

Figure 10 shows the absolute and percentage changes in  $PM_{2.5}$  concentrations under different temperature and humidity conditions in JJA of 2019 in 18 grids of BTH resulting from emission reductions. By reducing VOCs emissions, the highest and second largest values of  $\Delta PM_{2.5}$  concentrations occurred as T2 was above 32°C and was 471 26–28°C, respectively. With simultaneous reductions in NO<sub>x</sub> and VOCs emissions, the 472 absolute and percentage reductions in PM<sub>2.5</sub> mostly became larger as T2 increased. The 473 highest reductions in mean concentrations were 1.1 (3.5%), 1.0 (3.3%), and 0.9  $\mu$ g m<sup>-3</sup> 474 (3.0%) in the bin of >32 °C in cases of NO<sub>x</sub>\_10+VOCs\_10, NO<sub>x</sub>\_10+VOCs\_30, and 475 NO<sub>x</sub>\_10+VOCs\_50 simulations, respectively.

Reducing 10% of NO<sub>x</sub> emissions with 10%, 30%, and 50% of VOCs emissions were 476 simulated to result in the highest reductions of 1.6, 1.4, and 1.2  $\mu$ g m<sup>-3</sup>, respectively, in 477 478 mean concentrations of  $PM_{2.5}$  in the bin of 55–60%. The simulated largest percentage 479 decreases in mean  $PM_{2.5}$  concentrations occurred as RH was above 60%, with values of  $NO_x$  10+VOCs 10,  $NO_x$  10+VOCs 30, 480 3.7%, 3.2%, and 2.8% in and 481  $NO_x$  10+VOCs 50 simulations, respectively.



**483** Fig. 10. Same as Fig. 8, but for  $PM_{2.5}$ . The unit of absolute change is  $\mu g m^{-3}$ .

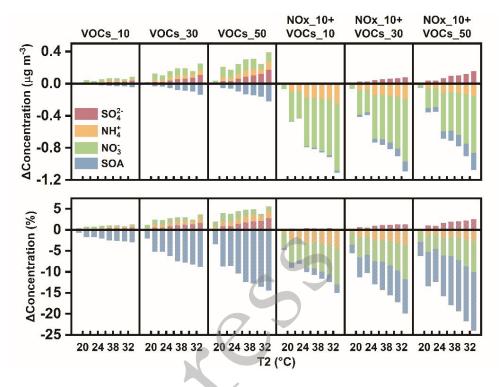
It was obviously shown that simultaneous reductions in NO<sub>x</sub> and VOCs emissions could alleviate  $PM_{2.5}$  pollution, but reductions in VOCs emissions alone exacerbated  $PM_{2.5}$  pollution. Reducing a smaller proportion of VOCs emissions resulted in a smaller increase in  $PM_{2.5}$  concentrations in cases of reducing VOCs emissions alone and a larger decrease in cases of simultaneously reducing NO<sub>x</sub> and VOCs emissions. As RH was above 55%, the control measures on VOCs emissions were simulated to lead to the highest decreases in  $PM_{2.5}$ .

### 491 *3.4.4 Impact of precursor emission reductions on PM*<sub>2.5</sub> *component concentrations*

The absolute and percentage changes in secondary components of PM<sub>2.5</sub> in each T2
and RH bins are presented in Figs. 11 and 12, respectively.

494 3.4.4.1 SOA

All of the six emission reduction measures led to reductions in SOA concentrations. 495 496 As T2 increased, the reductions in mean SOA concentrations became larger, consistent 497 with the reductions in O<sub>3</sub> and NO<sub>3</sub> concentrations (Figs. 7b-c), with the highest reductions of 4.6 × 10<sup>-2</sup> (3.0%), 0.1 (8.8%), 0.2 (14.5%),  $3.3 \times 10^{-2}$  (2.1%), 0.1 (8.1%), 498 and 0.2  $\mu$ g m<sup>-3</sup> (14.0%) in simulations of VOCs\_10, VOCs\_30, VOCs\_50, 499 500 NO<sub>x</sub> 10+VOCs 10, NO<sub>x</sub> 10+VOCs\_30, and NO<sub>x</sub>\_10+VOCs\_50, respectively. With respect to the results in RH intervals, the maximum absolute (percentage) decreases in 501 mean SOA concentrations of  $2.9 \times 10^{-2}$ – $0.2 \ \mu g \ m^{-3}$  (2.1%–15.8%) occurred at RH of 502 503 55–60% (above 60%), when the average concentrations of  $O_3$  had the highest (second highest) declines (Fig. 7f). Compared to the cases with reductions in both NO<sub>x</sub> and VOCs 504 505 emissions, reducing VOCs emissions alone resulted in greater reductions in SOA



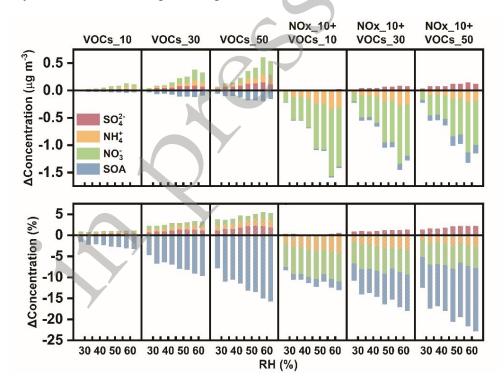
**Fig. 11.** Absolute (units:  $\mu$ g m<sup>-3</sup>) and percentage changes (units: %) in concentrations of PM<sub>2.5</sub> secondary components (SO<sub>4</sub><sup>2-7</sup>, NO<sub>3</sub><sup>-7</sup>, NH<sub>4</sub><sup>+</sup>, and SOA) in each T2 bin in 18 grids of BTH in JJA of 2019 (620 valid samples) in VOCs\_10, VOCs\_30, VOCs\_50, NO<sub>x</sub>\_10+VOCs\_10, NO<sub>x</sub>\_10+VOCs\_30, and NO<sub>x</sub>\_10+VOCs\_50 simulations (from left to right) relative to the CTRL simulation.

514 3.4.4.2 Sulfate

508

In contrast to SOA, six emission reduction measures all promoted the increases in SO<sub>4</sub><sup>2-</sup>. Controlling the emissions of NO<sub>x</sub> and VOCs led to the increases in OH radical (Figs. 7a and 7e), which enhanced the gas-phase oxidation of SO<sub>2</sub> and resulted in the increases in SO<sub>4</sub><sup>2-</sup>. Both the absolute and percentage increases of SO<sub>4</sub><sup>2-</sup> concentrations gradually increased as T2 went up, with the maximum of  $3.6 \times 10^{-2}$ –0.2 µg m<sup>-3</sup> and

0.6%–2.7%, respectively, except for the NO<sub>x</sub> 10+VOCs\_10 simulation. In the five cases 520 mentioned above, the positive changes of SOR were highest as T2 was above 32°C (Fig. 521 522 7d). In the NO<sub>x</sub> 10+VOCs 10 simulation, the highest absolute (percentage) increases 523 occurred as T2 was 26-28°C (24-26°C), when the increase in OH radical was second highest (highest) (Fig. 7a). In cases with reductions of VOCs emissions alone (both NO<sub>x</sub> 524 and VOCs emissions), the highest absolute and percentage increases in increased  $SO_4^{2-1}$ 525 concentrations occurred at RH of 50-60% (>55%), consistent with the changes in SOR 526 (Fig. 7h). In each T2 and RH bins, the absolute (percentage) increases in  $SO_4^{2-}$  were 527 528 positively correlated with the percentage reduction of VOCs emissions.



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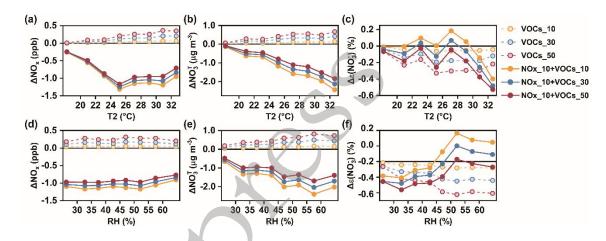
530 Fig. 12. Same as Fig. 11, but for each RH bin.

# 531 3.4.4.3 Nitrate

532 In the simulations with reductions of VOCs emissions alone, the higher proportion of 533 VOCs reduction, the larger the increase in  $NO_3^-$  concentration. The increases of OH 534 radical promoted the formation of  $HNO_3$  during daytime, while the formation of  $HNO_3$  at nighttime through hydrolysis was reduced due to lower N<sub>2</sub>O<sub>5</sub> from the decreases in O<sub>3</sub>. 535 The daily concentrations of  $NO_3^T$  increased as shown in Figs. 13b and 13e. Sulfate and 536 nitrate compete for NH<sub>3</sub>, so the enhanced  $SO_4^{2-}$  formation consumed more NH<sub>3</sub>, 537 resulting in a decrease in the proportion allocated to the particle phase in  $NO_3^T$  (as shown 538 in nitrate particle fraction ( $\epsilon(NO_3) = n - NO_3 / (n - HNO_3 + n - NO_3)$ ) in Figs. 13c and 539 13f). However, since  $NO_3^T$  was increased, there was still slight increase in daily 540 concentration of  $NO_3$ . Average over each T2 bin, the highest absolute (percentage) 541 increases in NO<sub>3</sub> concentrations occurred as T2 was 26-28°C (20-24°C), with values of 542  $3.2 \times 10^{-2}$  (0.4%),  $9.1 \times 10^{-2}$  (1.2%), and 0.1 µg m<sup>-3</sup> (2.0%) in VOCs 10, VOCs 30, 543 VOCs 50 simulations relative to the CTRL simulation, respectively. As T2 was 20-22°C 544 and 22-24°C, the increases in OH radical were highest and second highest, respectively 545 (Fig. 7a), leading to large formation of HNO<sub>3</sub> and  $NO_3^-$ . As for each RH interval, the 546 highest absolute (percentage) increases occurred when RH was 55–60% (above 60%). 547 with values of  $6.8 \times 10^{-2}$ – $0.3 \ \mu g \ m^{-3} (0.4\% - 1.7\%)$ . 548

With simultaneous reductions in NO<sub>x</sub> and VOCs emissions, the daytime and 549 nighttime formation of HNO<sub>3</sub> both slowed down due to the reductions in NO<sub>x</sub> and O<sub>3</sub> 550 concentrations, inhibiting the generation of  $NO_3^T$  and leading to the decreases in daily 551 552 concentrations of  $NO_3^{-1}$ . The smaller proportion of VOCs reduction resulted in the larger decreases in NO<sub>3</sub> concentrations due to the smaller increases in OH (Figs. 7a and 7e) 553 and the larger decreases in  $NO_x$  (Figs. 13a and 13d), which inhibited both daytime and 554 nighttime HNO<sub>3</sub> formation more significantly. As T2 went up, the reductions in mean 555  $NO_3$  concentration exhibited higher efficiency (larger reductions), consistent with the 556

557 reductions in O<sub>3</sub> and NO<sub>3</sub> concentrations (Figs. 7b-c). The highest reductions were simulated to be 0.8 (8.5%), 0.8 (8.1%), 0.7  $\mu$ g m<sup>-3</sup> (7.3%) in simulations of 558 NO<sub>x</sub> 10+VOCs 10, NO<sub>x</sub> 10+VOCs 30, and NO<sub>x</sub> 10+VOCs 50, respectively. Average 559 over each RH bin, the highest reductions in  $NO_3^-$  concentrations occurred when RH was 560 561 55–60% (the RH bin with the largest decrease in mean O<sub>3</sub> concentration), with values of 1.1, and 0.9  $\mu$ g m<sup>-3</sup> in NO<sub>x</sub> 10+VOCs 10, NO<sub>x</sub> 10+VOCs 30, 562 1.2. and 563  $NO_x$  10+VOCs 50 simulations relative to the CTRL simulation, respectively.



**Fig. 13.** Same as Fig. 7, but for surface-layer (a/d) NO<sub>x</sub> (units: ppb), (b/e) NO<sub>3</sub><sup>T</sup> (units:  $\mu g m^{-3}$ ), and (c/f) nitrate particle fraction ( $\epsilon(NO_3^-) = n-NO_3^- / (n-HNO_3 + n-NO_3^-)$ ; units: 567 %).

568 3.4.4.4 Ammonium

564

The reductions in emissions of VOCs and  $NO_x$  affect the concentrations of  $NH_4^+$ through the formation of  $SO_4^{2-}$  and  $NO_3^-$ . Reducing VOCs emissions alone ( $NO_x$  and VOCs emissions simultaneously) resulted in an increase (a decrease) in  $NH_4^+$ concentrations. As T2 increased, the increases (decreases) in mean  $NH_4^+$  concentrations became larger in both cases. The highest reductions in mean  $NH_4^+$  concentrations were 574 simulated to be 0.1–0.2  $\mu$ g m<sup>-3</sup> (2.8%–4.5%) with the simultaneous reductions in NO<sub>x</sub> 575 and VOCs emissions. For each RH interval, the highest absolute and percentage increases 576 (decreases) in mean NH<sub>4</sub><sup>+</sup> concentrations occurred when RH was 55–60% and was 577 above 60% in both cases, respectively.

578 Overall, simultaneously reducing  $NO_x$  and VOCs emissions alleviated  $PM_{2.5}$ pollution effectively, especially with a smaller proportion of VOCs reduction. With 579 580 simultaneous reductions in NO<sub>x</sub> and VOCs emissions, although there was a positive 581 correlation between the decrease in SOA concentrations and the percentage reduction of VOCs emissions, the impact of SOA changes on the level of PM<sub>2.5</sub> was weak due to its 582 small proportion in PM<sub>2.5</sub>. Smaller percentage reduction in VOCs resulted in the smaller 583 increase in  $SO_4^2$  concentrations due to the smaller increase in OH radicals, and led to 584 the higher decrease in  $NO_3^-$  concentration owing to the larger reduction in both daytime 585 and nighttime formation of HNO3 caused by the larger decrease in NOx. At higher 586 temperatures, simultaneous reductions in NO<sub>x</sub> and VOCs emissions generated more 587 sulfate but significantly suppressed the generation of SOA, nitrate, and ammonium due to 588 the larger decreases in O<sub>3</sub> and NO<sub>3</sub>, leading to larger reductions in PM<sub>2.5</sub> concentrations. 589 When RH was above 55%, the absolute and percentage decreases in  $PM_{2.5}$  concentrations 590 reached the maximum values, caused by the highest reductions in SOA,  $NO_3^-$ , and  $NH_4^+$ 591 592 concentrations due to the highest reduction in O<sub>3</sub> concentrations.

593 4. Conclusions

We used the observed hourly concentrations of  $O_3$  and  $PM_{2.5}$  from MEE, meteorological parameters from MERRA-2, and the model results from the nested-grid version of the GEOS-Chem model to examine the relationships between  $O_3$ ,  $PM_{2.5}$ , and 597  $PM_{2.5}$  components and T2/RH over the BTH and to investigate the effectiveness of 598 precursor emission reductions on O<sub>3</sub> and PM<sub>2.5</sub> in BTH under different temperature and 599 humidity conditions in the summer of 2019.

Both observed (simulated) MDA8 O<sub>3</sub> and PM<sub>2.5</sub> concentrations were found to 600 increase as T2 went up, with linear trends of 4.8 (3.2) ppb  $^{\circ}C^{-1}$  and 1.9 (1.5) µg m<sup>-3</sup>  $^{\circ}C^{-1}$ , 601 602 respectively. Atmospheric oxidation capacity was stronger at higher temperature, 603 resulting in the increases in PM<sub>2.5</sub> secondary components. As RH increased, concentrations of PM<sub>2.5</sub> secondary components increased first and then decreased. SOA 604 peaked at RH of 45–50% when  $O_x$  and NO<sub>3</sub> were at a high level, while  $SO_4^{2-}$ , NO<sub>3</sub>, and 605  $NH_4^+$  peaked at RH of 55–60%. There was a bimodal trend in observed/simulated 606 MDA8  $O_3$  concentrations (peaking at 30–35% and 45–50%/30–35% and 55–60%). Both 607 608 observed and simulated  $PM_{2.5}$  concentrations peaked at RH of 55–60%.

The results of the six sensitivity experiments showed that implementing precursor 609 610 emission reductions can effectively mitigate summer O<sub>3</sub> pollution in BTH. The larger the 611 percentage reductions of VOCs emissions, the larger the reductions in MDA8 O<sub>3</sub>. As T2 went up, the absolute values of  $\Delta$ MDA8 O<sub>3</sub> showed an overall increasing trend, with the 612 maximum values of 0.9–5.2 ppb at T2 of >32°C in the six simulations. The percentage 613 614 reductions in MDA8 O<sub>3</sub> were the largest (0.9%–4.9%) at T2 of 30–32°C with reductions in VOCs emissions alone and (1.7%-5.4%) at T2 of >32°C with simultaneous reductions 615 in NO<sub>x</sub> and VOCs. When RH increased, the decreases of MDA8 O<sub>3</sub> concentrations 616 exhibited a bimodal trend. Among the six simulations, the largest decreases in MDA8 O<sub>3</sub> 617 618 (4.6 ppb (5.4%)) occurred at RH of 55–60% with a 50% reduction in VOCs emissions.

619 Different from the responses of O<sub>3</sub>, reducing VOCs emissions alone exacerbated 620 PM<sub>2.5</sub> pollution. Simultaneous reductions in NO<sub>x</sub> and VOCs emissions alleviated summer 621 PM<sub>2.5</sub> pollution in BTH. Reducing a smaller proportion of VOCs resulted in a larger decrease in  $PM_{2.5}$  concentration, driven by the larger decrease in  $NO_3^-$  concentration due 622 623 to the larger reduction in both daytime and nighttime formation of HNO<sub>3</sub> from the larger decrease in NO<sub>x</sub>. As T2 increased, reductions in PM<sub>2.5</sub> mostly became larger, with the 624 625 peak values of 1.1 (3.5%), 1.0 (3.3%), and 0.9  $\mu$ g m<sup>-3</sup> (3.0%) at T2 of >32°C, respectively, in NO<sub>x</sub> 10+VOCs 10, NO<sub>x</sub> 10+VOCs 30, and NO<sub>x</sub> 10+VOCs 50 626 simulations. When T2 was above 32°C, the reductions in SOA, NO3, and NH4<sup>+</sup> 627 concentrations reached their maximum values owing to the large decreases in O<sub>3</sub> and 628 NO<sub>3</sub>. As for the RH intervals, the highest absolute reduction in PM<sub>2.5</sub> concentrations 629 630  $(1.2-1.6 \ \mu g \ m^{-3})$  occurred at RH of 55-60% and the highest percentage reduction (2.8%-3.7%) occurred in the bin of >60% with simultaneous reductions in NO<sub>x</sub> and 631 VOCs. At RH of >55%, the large reductions in O<sub>3</sub> contributed to the large reductions in 632 SOA,  $NO_3^-$ , and  $NH_4^+$  concentrations. 633

In summary, reducing a larger proportion of VOCs emissions can lead to larger reduction in MDA8  $O_3$  in BTH during summer. The decreases in MDA8  $O_3$  caused by precursor emission reductions are more sensitive to T2 than to RH. Higher reduction of VOCs emissions at higher T2 is more effective for alleviating summer  $O_3$  pollution in BTH. When PM<sub>2.5</sub> pollution is a concern in summer, NO<sub>x</sub> reduction combined with a small proportion reduction of VOCs is the best measure. The reduction in PM<sub>2.5</sub> from reducing precursor emissions is more sensitive to RH than to T2, with the best efficiency at high RH. For the control of co-pollution of  $O_3$  and  $PM_{2.5}$  in summer in BTH, precursor emission reductions have the best effect in a hot and humid conditions.

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